Solid ^{100}Mo Target Preparation Using Cold Rolling and Diffusion Bonding

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Introduction

With interruptions in reactor-based supply of ^{99}Mo/^{99m}Tc, there is growing interest in using cyclotrons for direct production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction. Although the yield of ^{99m}Tc is theoretically directly proportional to the beam current, the design and fabrication of enriched ^{100}Mo targets is one of the challenges that had previously limited the large-scale production.

Several ^{100}Mo target design considerations are required. The target back plate supporting the ^{100}Mo must be chemically inert to target dissolution conditions but ideally it should also be able to dissipate high thermal loads of irradiation, not contaminate target substrate with radionuclidic by-products, and be adequately inexpensive to allow for single use. Aluminum was selected as a target support because it is a reasonable balance of these requirements.

The process we have developed for ^{100}Mo target fabrication entails rolling ^{100}Mo powder into a foil of desired thickness, and then using diffusion bonding [1] to bond the foil onto an aluminum back plate. The ^{100}Mo targets were designed to be approximately 20 mm by 80 mm by 0.1 mm to match the ACSI TR 24 cyclotron's beam profile and energy.

Material and Methods

With the full cycle noted in Figure 1, the crude enriched^{100}Mo foils (99.815% enrichment) were made from rolling ^{100}Mo powder using a horizontally mounted rolling mill and an aluminum hopper. The crude foils were rolled repeatedly, and the space between the rollers gradually reduced until the thickness of the foils changed from an initial thickness of 0.3 mm to a thickness of 0.1 mm. The foils were then trimmed to achieve the 80 mm length.

The rolled ^{100}Mo foils (n=20) were annealed in a 5.11% hydrogen in argon atmosphere for 1 hour at 1300°C (temperature ramped at 2°C/min until 75°C and then 5°C /min until 1300°C). The surface area of the annealed foils were measured using Image J software and the mass and density of the foils were measured using a Sartorius YDK03 density kit on a Sartorius analytical balance.

The surface of the 6061 series aluminum back plates and annealed ^{100}Mo foils were cleaned with 180, 320 and finally 400 grit sandpaper. The annealed ^{100}Mo foils were bonded to the aluminum back plate in a pre-heated press at 500°C with a 2200 kg load on the platens of the press. Targets were then screwed onto a target support base in preparation for irradiation. When docked in the target station, targets are oriented at 7 degree angle to the proton beam, the target face is open to vacuum, and the target is cooled with ~40 L/min chilled water. Targets are automatically loaded/retrieved by means of a pneumatic target transfer system.

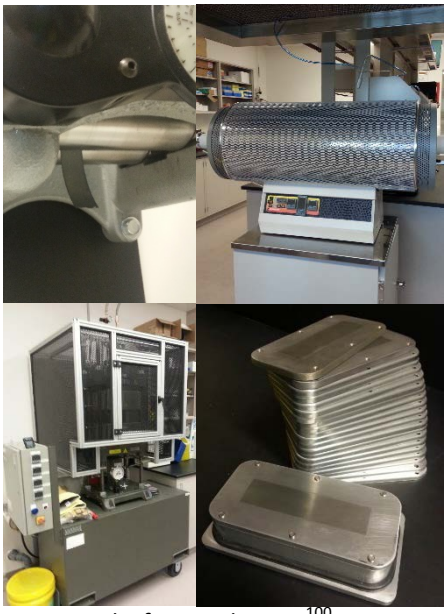

FIGURE 1. Cycle for producing ^{100}Mo targets including rolling of powder, annealing under reducing atmosphere, and diffusion bonding by means of a heated press.

Prior to ^{100}Mo irradiation, and for purpose of minimizing beam tuning time, irradiations were performed on blank aluminum "dummy" targets to optimize the cyclotron injection system and beamline parameters. Irradiations on these aluminum-only targets were repeated until such point that we were satisfied with the beam profile as indirectly measured by means of Gafchromic film [2].

A series of eight ^{100}Mo target irradiations were then performed at a nominal proton energy of 22 MeV. The first target was irradiated at

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150 µA for 15 minutes (excluding tuning time). Each subsequent irradiation was successively increased by 50 µA until 500 µA was achieved. Tuning time was typically less than 5 minutes, and the total integrated current on target was recorded for purpose of yield calculation.

Irradiation current "on" target was measured by physical addition of the current measurements on the target and the electron catcher, biased at 45 V. Beam "spill" was calculated and is defined here as the combined current on the collimators and mask, divided by the sum of currents on the collimators, mask, and biased target/electron catcher. Beam spill for the eight irradiations was 26 ± 4%.

For purpose of determining ^{99m}Tc yield, we opted not to chemically process the targets so as to minimize any losses due to chemistry inefficiencies. Target plates were therefore removed from the target support and directly placed in a dose calibrator. Multiple measurements and curve fitting were employed to extrapolate the ^{99m}Tc contribution to the measurement. Self-attenuation through the ^{100}Mo was not accounted for.

Results and Conclusion

The 20 targets produced by cold rolling and diffusion bonding showed that high density targets, >96% compared to the maximum theoretical density of ^{100}Mo, with the physical properties required for the beam profile were able to be produced as shown in Table 1.

97% of the ^{100}Mo powder was recovered during the target preparation or made into a ^{100}Mo target. All twenty of the prepared ^{100}Mo targets demonstrated good bonding to the aluminum substrate by visual inspection.

Mass [g]	2.05 ± 0.16
Aerial density [mg/cm^2]	135 ± 11
Density [g/cm^3]	10.26 ± 0.07
Thickness [µm]	131 ± 11
Area [cm^2]	15.3 ± 0.09

TABLE 1. Average and standard deviation of ^{100}Mo target physical properties (n=20).

The 8 irradiated targets were completely intact after the 15 minute irradiation with the exceptions of the targets irradiated at 350 µA and 500 µA. These exceptions showed burn marks on the surface as shown in Figure 2.

The burn marks are attributed to areas of poor bonding between the ^{100}Mo foil and the aluminum back plate which would cause poor thermal conductivity. Based on spikes in the vacuum, however, we believe these burns occurred early during the tuning, and not as result of reaching the desired 350 or 500 µA beam currents.

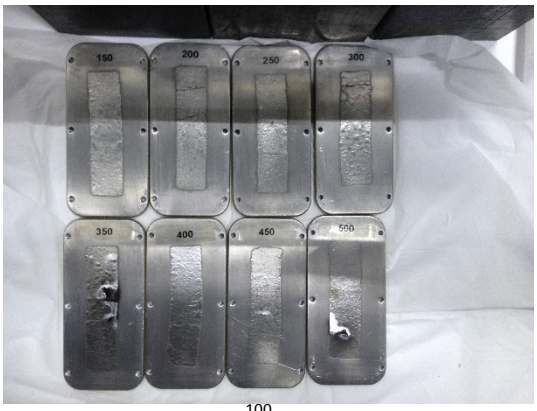

FIGURE 2. Irradiated ^{100}Mo targets at varying beam currents

Despite the burn marks on the 350 µA and 500 µA targets, high yields of ^{99m}Tc activity were achieved as shown in Figure 3. In comparison to the expected thick target theoretical yield of 6.79 GBq/µA as calculated from [3], an average yield of 6.1 ± 0.2 GBq/µA was obtained (or 89 ± 3% of expected). There was also no obvious trending towards a decrease in yield as the irradiation current was increased.

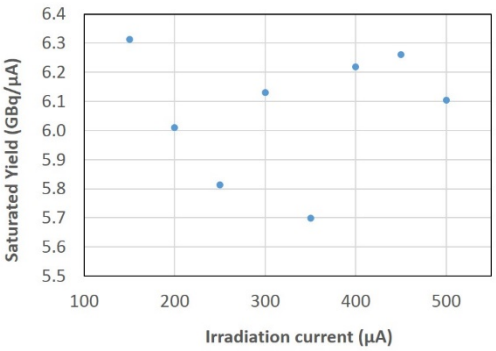

FIGURE 3. Experimental saturated yields for irradiated ^{100}Mo targets as a function of irradiation current.

The ^{100}Mo targets prepared for ^{99m}Tc production were relatively robust. The 350 µA and 500 µA targets with burn marks still had 84% and 89% of their expected yields respectively. The other 6 targets showed no burn marks from the irradiations. No correlation between burn marks and loss of yield were noted.

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No targets in these experiments were chemically processed. Previous experiments with targets made from molybdenum with natural isotopic abundance and this method of target preparation have been chemically processed with targets in under 30 minutes using 30% H_2O_2.

In considering large-scale production, the irradiation at 500 μA demonstrated production of >100 GBq (>3 Ci) of ^{99m}Tc in approximately twenty minutes. Extrapolation of this yield to longer irradiation times suggests we can produce ~1.5 TBq (41 Ci) in 6 hours. Irradiations at 1, 3, and 6 hours at 500 μA are planned for purposes of confirming these yields.

We are also looking to identify a pre-irradiation QC method for the target plates which would ensure uniform bonding across the entire Mo/Al interface. Future studies will require pressing multiple targets at once and ultimately, automation of the cold rolling and diffusion bonding process to increase the rate at which the targets can be produced to further lower the production costs of the ^{100}Mo target.

These results demonstrate a robust, relatively inexpensive target with desirable physical properties that can handle high irradiation currents during large scale production of ^{99m}Tc via the ^{100}Mo(p,2n)^{99m}Tc reaction.

References

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